
Electrode Architecture-Assembly of Battery Materials and Electrodes

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DOE-BATT Review Meeting

ID # ES222

Overview

Timeline

- Start date: March 2013
- End date: December 2016
- 35% completed

Barriers

- Low energy
- Poor cycle/calendar life

Budget

- Total Project Funding: \$2.92M

	DOE	HQ
FY13 funding	\$365K	\$365K
FY14 funding	\$365K	\$365K
FY15 funding	\$365K	\$365K
FY16 funding	\$365K	\$365K

Partners

- V. Battaglia, V. Srinivasan and R. Kosteckı (LBNL)
- J. Goodenough (U. Texas)
- C. Julien, A. Mauger (U. Paris 6)
- X.Q. Yang (BNL)

Objectives

- **Develop high-capacity, low-cost electrodes with good cycle stability and rate capability.**
- **Identify a low-cost method to produce Si nano-powder.**
- **Identify the mechanism of electrode degradation by using *in-situ* tools to improve the electrode composition and architecture.**

Approach

- **Design of electrode architecture by controlling tortuosity and porosity and maintaining high ionic conductivity.**
- **Develop an improved electrode architecture of Si nano-particles with carbon and water-based binders.**
- **Metallurgical Si is used to produce a low cost Si nano-powder.**
- **Utilize *in-situ* and *ex-situ* SEM and TEM to investigate the SEI layer on the anode and cathode.**

Milestones

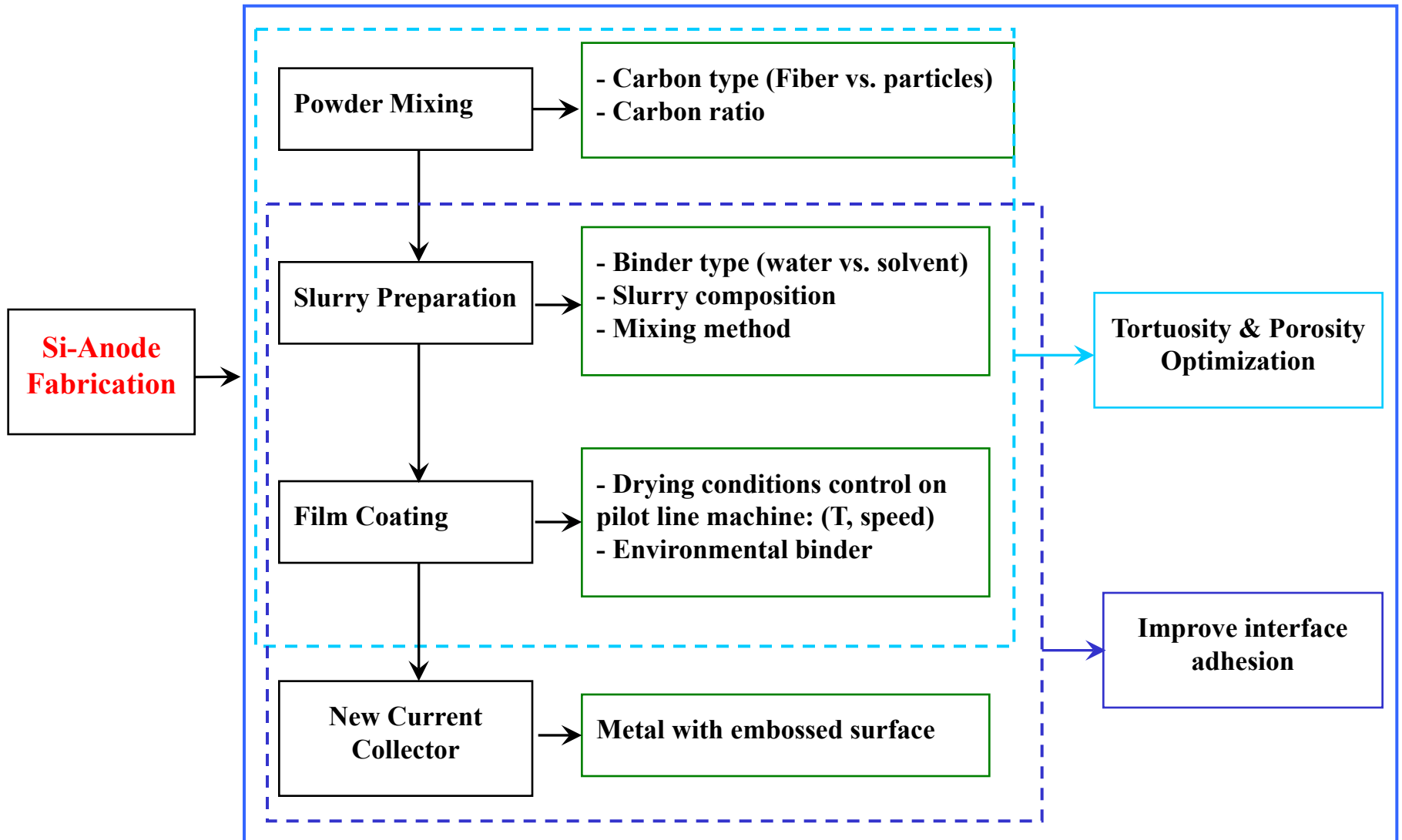
■ Completed:

- Identify Si-based anode materials that can achieve a capacity of 1200 mAh/g.
- Supply Si powder (1 Kg) from an alternative supplier as a baseline material for BATT PIs.

■ On going:

- Go/No-Go: Terminate production of Si powder in anode tests that show more than 20% capacity fade in the initial 100 cycles.
- Criteria: Supply laminates Si-based electrodes to BATT PIs.
- Supply a 20-Ah Li-ion flat cell based on Si and LiMNO materials to BATT PIs.

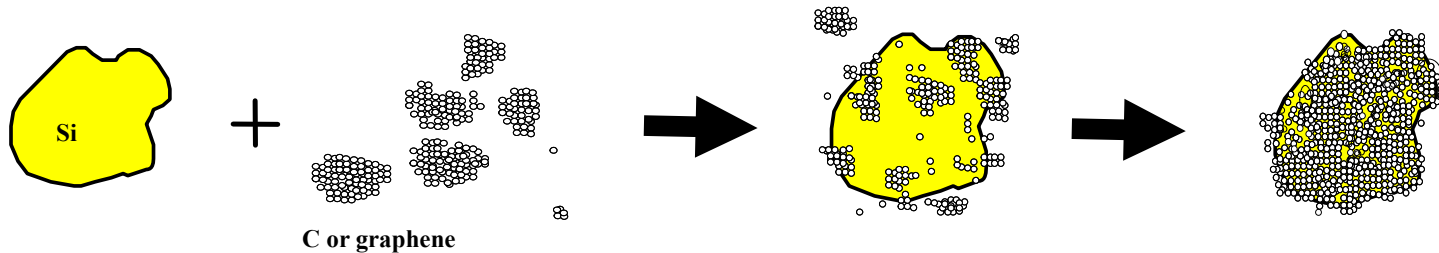
Plan to Achieve Milestones



Powder Dispersion

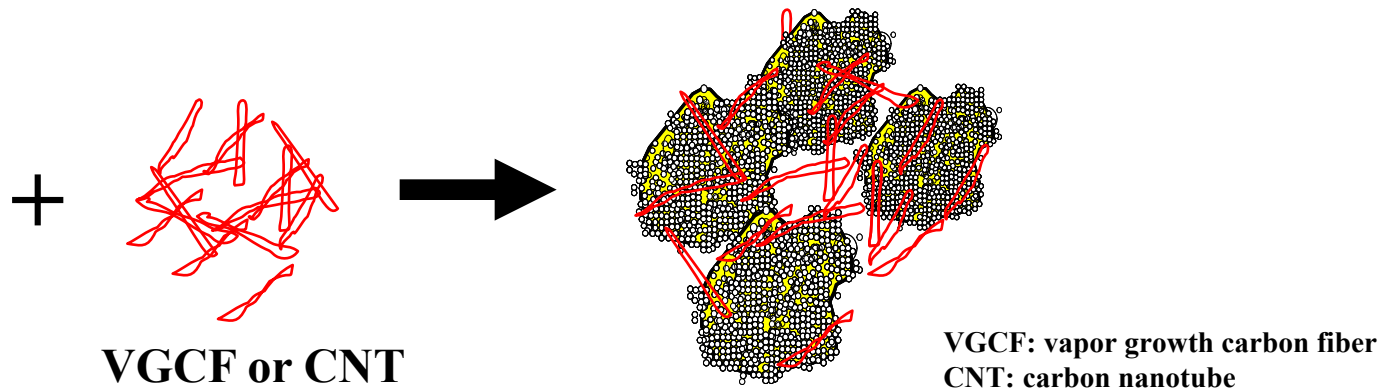
Step1

Premix step prevents the agglomeration (macrophase separation) of carbon and active materials.

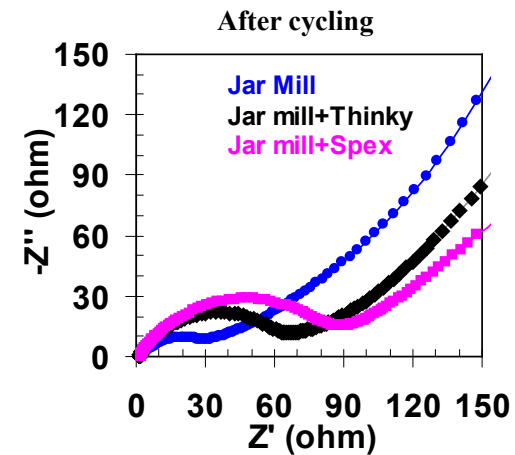
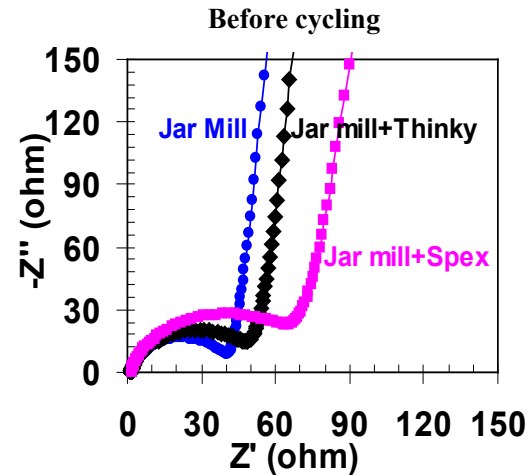
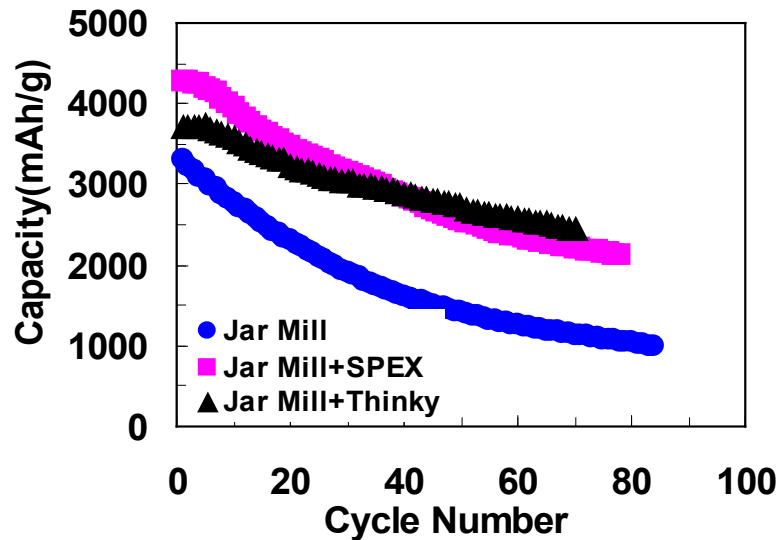


Step2

Then, carbon fibers will be added to improve the electric contact between Si particles even after volume expansion.



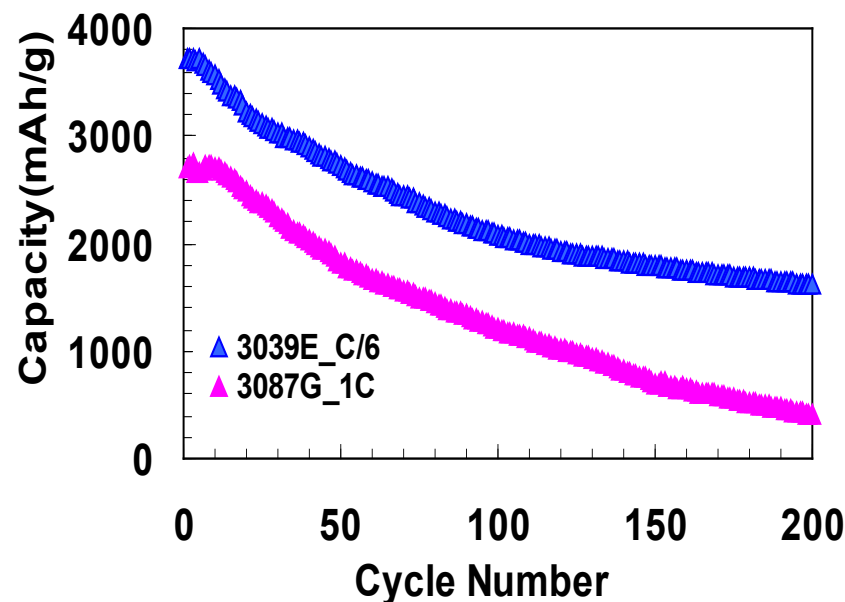
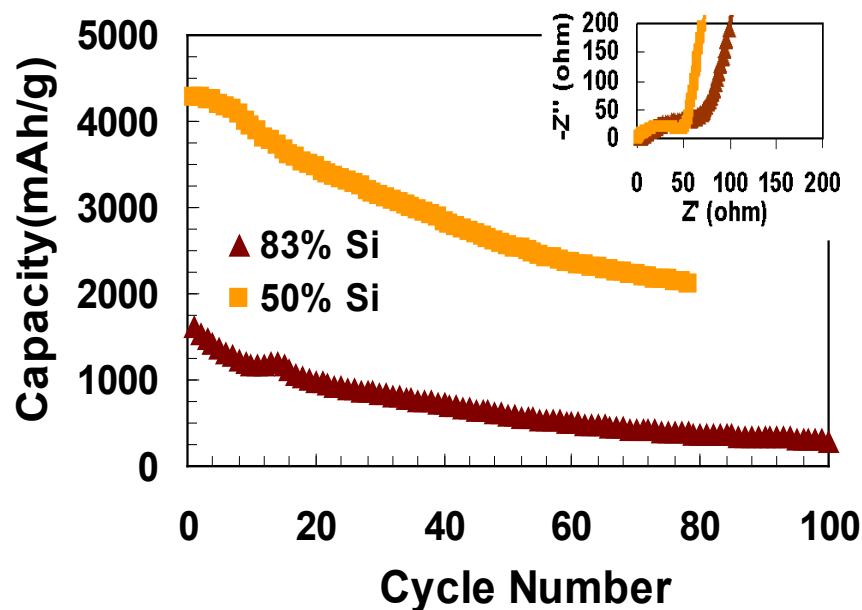
Effect of Mixing Method on Cycling & Impedance



Mixing methods	Reversible cap. (mAh/g)	Cap. fade After 70 cycles (%)
Jar mill	3300	65
Jar mill + Ball mill	4280	48
Jar mill + Planetary mix	3710	34

The jar-mill alone is not effective as a mixing method.

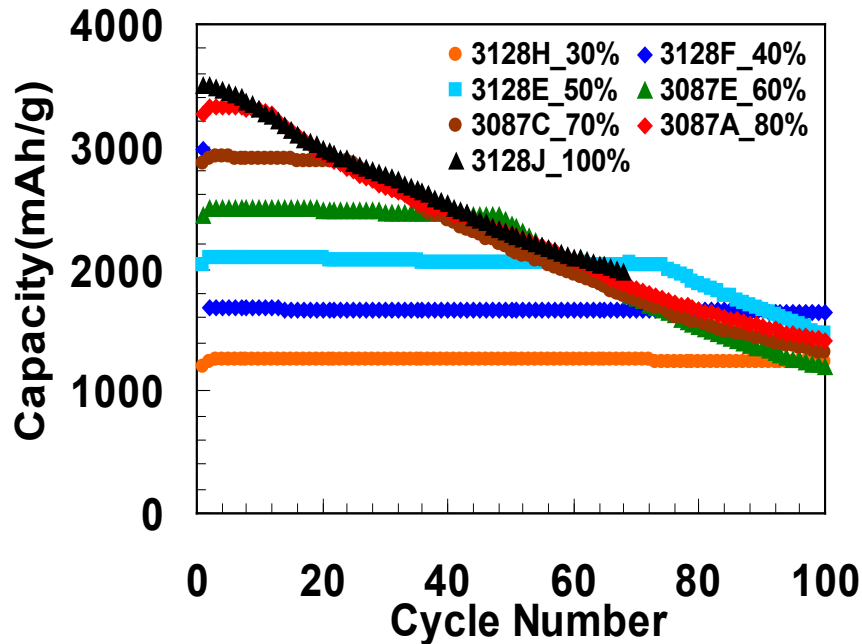
Effect of Loading & Rate on Cycling



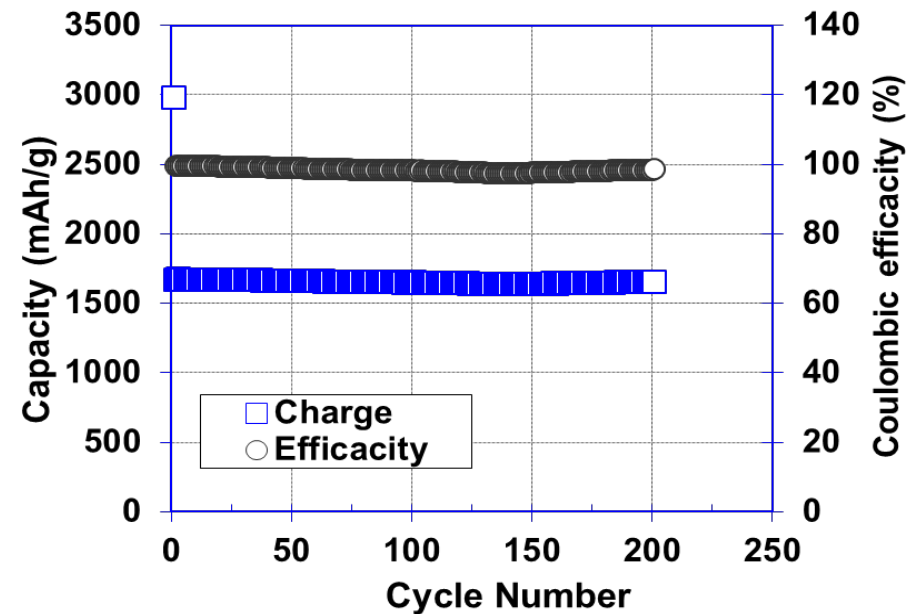
- High loading affects the performance by inducing a large accumulation of stress in the anode.
- Carbon improved electric contact between particles, even after cracking.

The Si particle suffers more at high rate (1C), which induces a fade of 85% of the initial capacity compared to 56% at C/6.

Effect of Depth of Discharge on Cycling

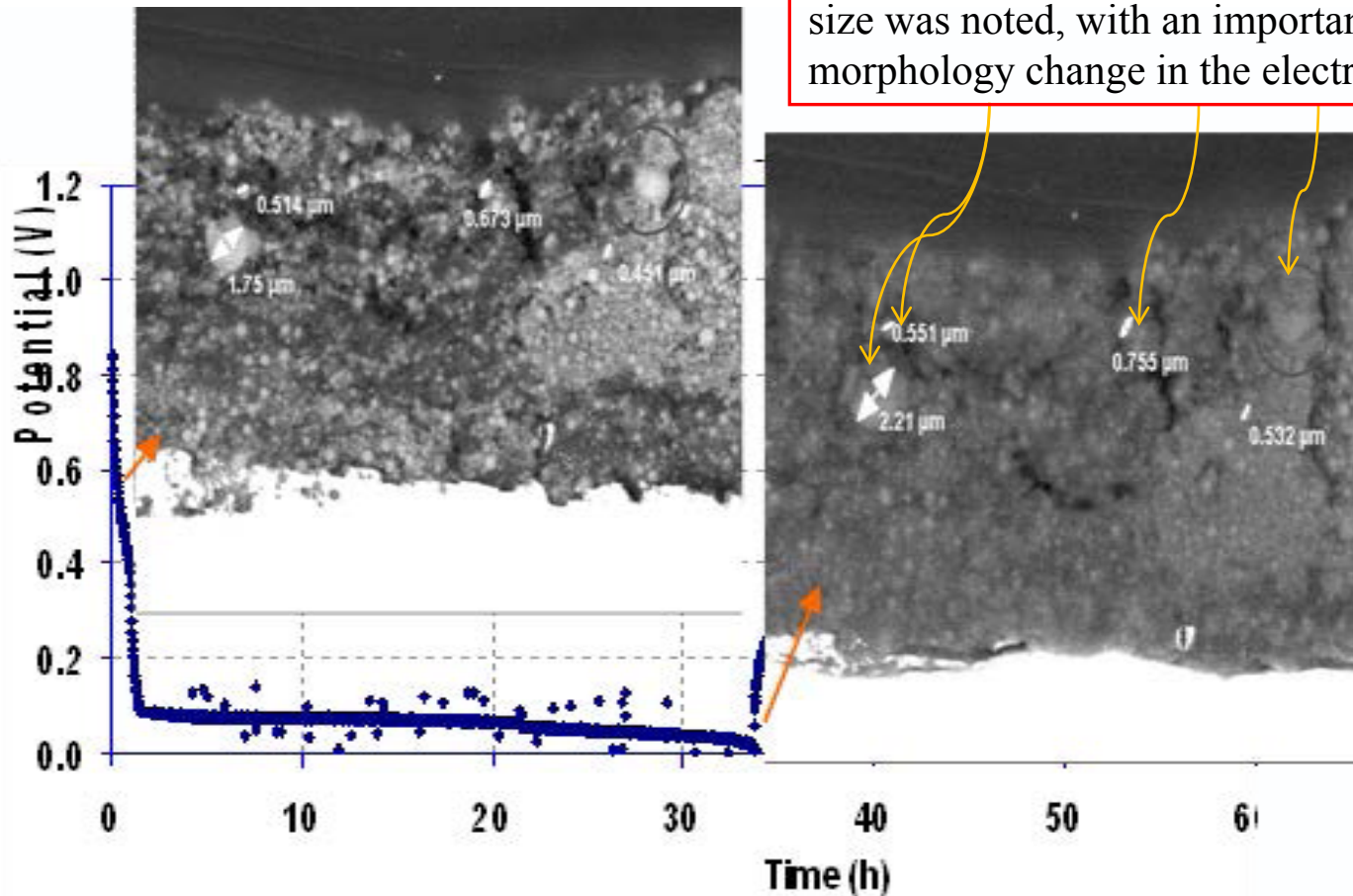


The stress level was controlled by varying DoD, at low DoD, the Si particles in the anode will experience minimum stress.



At 40% DoD, a very stable reversible capacity of around 1670 mAh/g with very good coulombic efficiency is obtained.

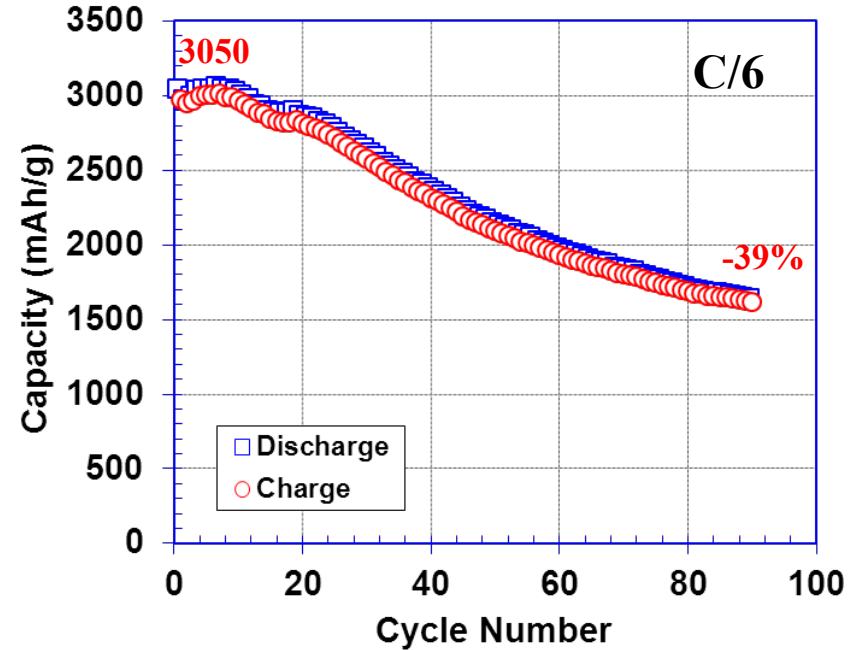
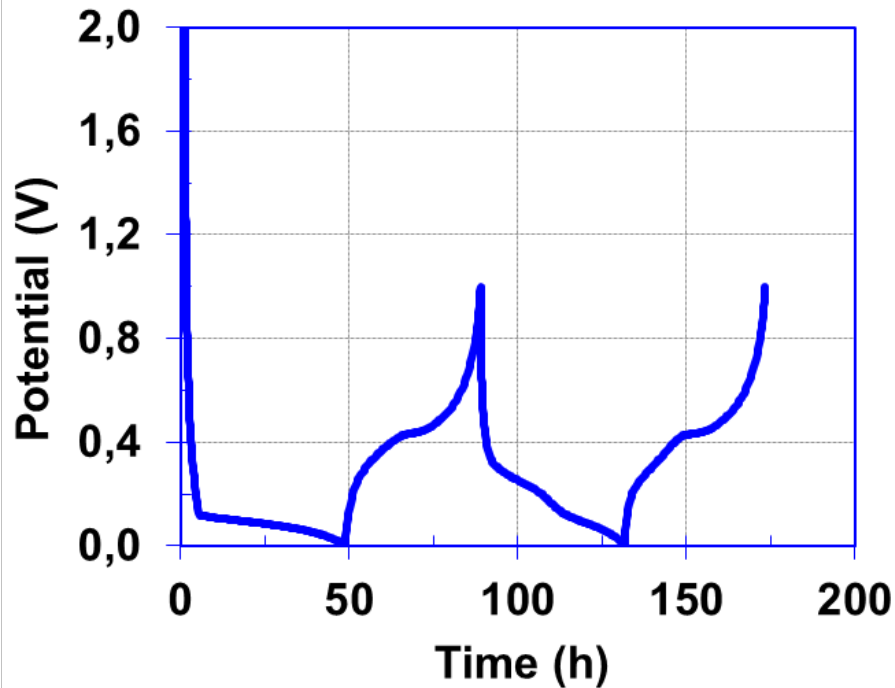
In-Situ SEM Analysis



An increase in the primary particle size was noted, with an important morphology change in the electrode.

The results of the *in-situ* analysis revealed an increase in the electrode thickness during discharge to 5 mV, and the average electrode expansion was about 58%.

HQ Si-nano Characterization



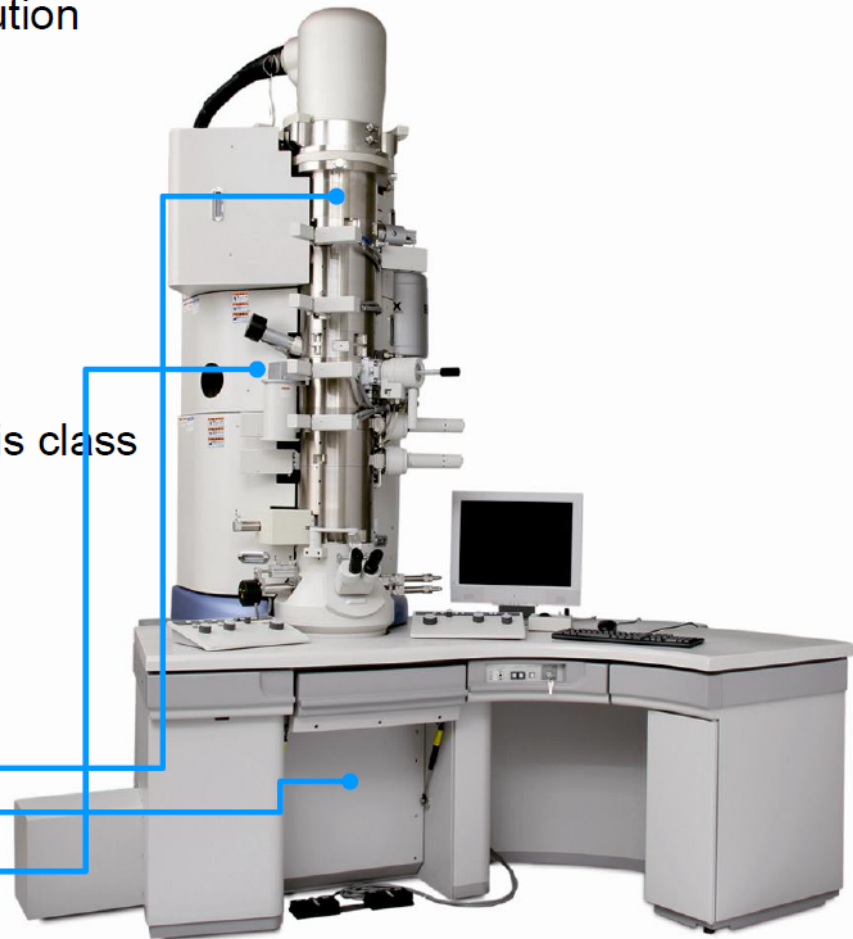
- High reversible capacity was obtained with Hydro-Quebec nano-Si with 3050 mAh/g and high efficiency (~100%).
- Cycling at C/6, Si-HQ shows a capacity fade of 39%.

New TEM Microscope (HF-3300)

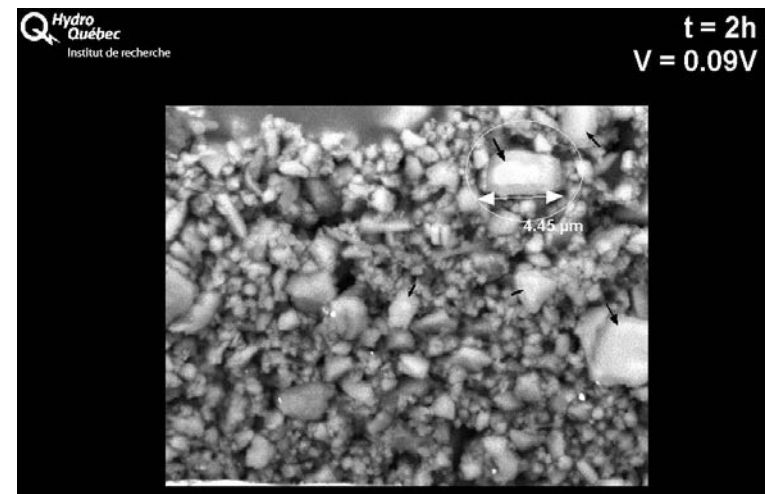
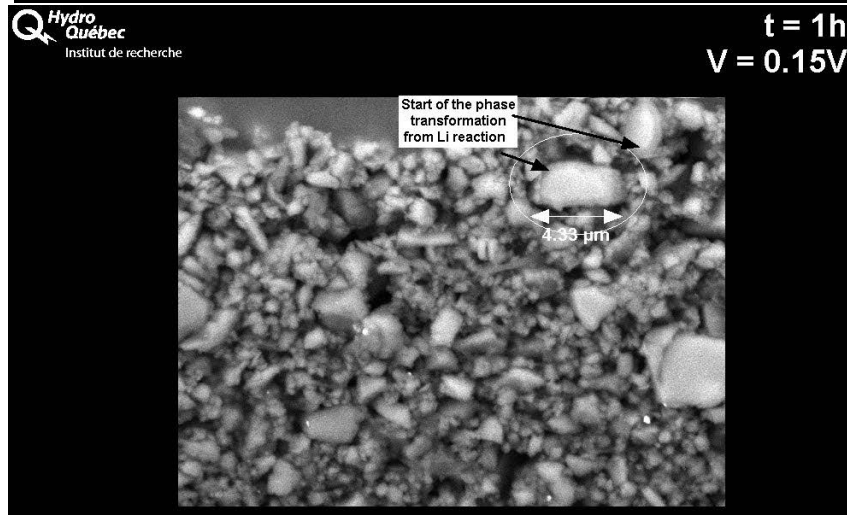
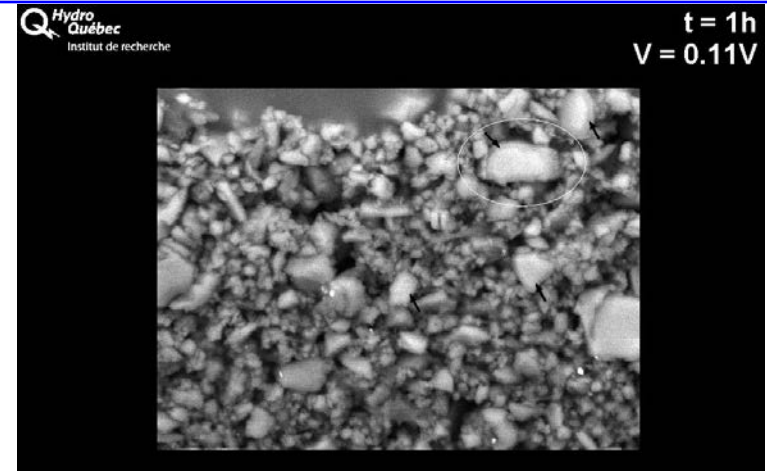
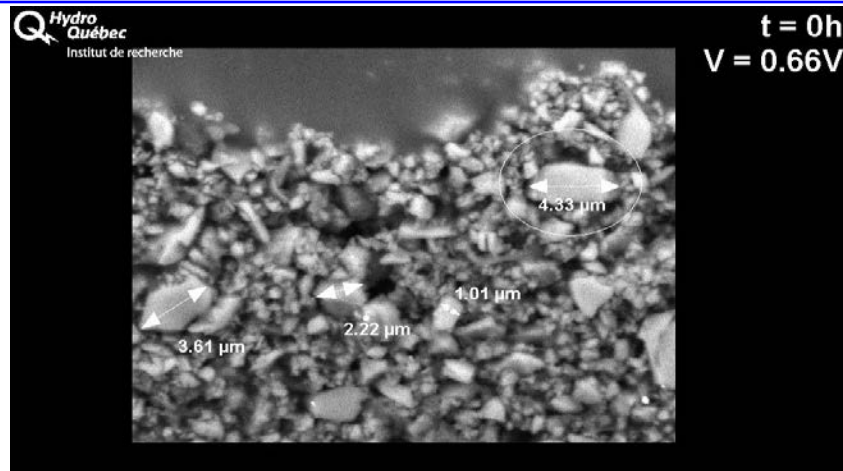
Features

- ▶ New Cold field emission gun
 - High brightness & high energy resolution
 - Low tip noise and emission decay
 - Long life
- ▶ High performance of FE-TEM/STEM
 - High contrast/resolution
 - Low voltage capability*
- ▶ High sensitive EDX analysis*
 - The widest solid angle (0.24 sr) in this class
- ▶ EELS performance*
 - High energy resolution (<0.5 eV)
 - Chemical analysis (<0.2 eV)
- ▶ Advanced analytical approach*
 - Nano beam diffraction
 - Spatially resolved EELS
 - **In-situ observation**

*Option



In-Situ SEM: 1st Discharge of Si-Based Anode



- *In-situ* SEM shows the volume expansion of several Si-based particles.
- No evidence of cracking was observed with any of the Si-particle morphologies.

Summary

- Improved silicon anode was developed that utilized nano-Si and water-based binder (alginate).
- A high-carbon content and low-Si ratio in the anode composition yielded a significantly better performance.
- The stress on the Si particles was limited by controlling depth of discharge (DoD); when the anode is cycled at a deep DoD, the capacity fade is severe, which is believed to be due to high stress and cracking.
- This result indicates that sacrificing some capacity may be an acceptable compromise for achieving longer cycle life while maintaining adequate capacity.
- The results of the *in-situ* analysis revealed an increase in the electrode thickness during discharge to 5 mV, and the average electrode expansion was about 58%.
- The volume expansion was dependent on the electrode composition.

Future Activities

☐ Remainder of this year

- Produce 10 m length of Si-anode by using the HQ pilot coating machine for BATT PIs.
- Fabricate 20Ah Li-ion stacking battery by using Si-anode and LiMNO.
- Increase the Si-loading in the anode.
- Study the DoD effect on the performance of the 20 Ah Li-ion battery.



Pilot line stacking machine @ IREQ